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Quantifying Atmospheric Deposition of Phosphorus: A Conceptual Model and Literature Review for Environmental Management

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Executive Summary

This paper describes concepts underlying the atmospheric input of phosphorus (P) to ecosystems, summarizes published rates of P deposition and outlines improvements for estimating rates of P deposition. Attempts to evaluate atmospheric contributions of P to ecosystems have proven problematic due to limitations of particle sampling methods, lack of a means for validating total P deposition rates, and persistent sample contamination. Particles conveying P are primarily greater than 2 μm (microns) in diameter, about the size of a bacterial cell, and increase greatly in abundance with land disturbances associated with urbanization and agriculture. Coarse particles (>10 μm) can be important agent moving P in spirals of retention and transport across the landscape, and represent the greatest measurement challenge due to high settling velocities. Plant canopies can alter P deposition rates greatly across landscapes through retention and release or generation of P-bearing particles.

Published annual rates of P deposition range from about 5 to well over 100 mg P/m^2/yr, although most values fall between 20 and 80 mg P/m^2/yr. Rates below 30 mg P/m^2/yr are found in remote areas and near coastlines; immediate rates of 30 to 50 mg P/m^2/yr are associated with forests or mixed land uses; and rates of 50 or more are recorded from urban or agricultural settings.

The rate of wet deposition of P from the atmosphere can be quantified using common bucket collectors with careful attention to sample processing and site selection to minimize contamination. For dry P deposition, a review of available methods revealed that a monitoring program should estimate inputs for the size classes of particles carrying airborne P and use an inferential approach measuring atmospheric concentrations and deposition velocities. Inferential rates should be applicable to the landscape level to account for canopy effects and gradients in particle concentrations across an aquatic environment. Separating natural organic inputs of P from sample contamination will continue to be challenging. Calibrated surface accumulation methods will be required to sample intermediate and coarse-sized particles to provide direct measurements of total particle flux by size class. Atmospheric P concentrations can be obtained with high-volume (ca., 1.1 m^3/min) particulate matter samplers. Sampling stations should be located for upwind/downwind gradients to allow *in situ* estimation of differences in concentration and net deposition velocities. Improved statistical tools and methods for detecting biological contamination are needed for data screening. The information summarized in this study should encourage more routine measurements of P deposition rates in national atmospheric monitoring programs.

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Introduction

The flux or loading of nutrients to ecosystems occurs primarily through point source discharges, surface water, ground water and atmospheric deposition. The input of nutrients from wastewater discharges has become less of a problem over the last one-quarter century and can be quantified from discharge monitoring records. Loading via surface water has been measured for many ecosystems and can be the major source of nutrients for aquatic environments, particularly those with large watersheds. Ground water inputs are more difficult to assess and are often estimated indirectly by mass balance calculations. Nutrients associated with rainfall and dry fallout from the atmosphere have been found to be an important source of nutrients to many freshwater environments (Axler et al., 1994; Gorham, 1961; Jassby et al., 1994; Lewis, 1983). Graham and Duce (1982) estimated that phosphorus deposition from the atmosphere to the world's oceans represents about 10% of riverine inputs to the oceans from continents. Conversely, the oceans may contribute substantial amounts of P to continents through aerosols injected into the air from the ocean surface (Graham et al., 1979). Rigorous estimates of atmospheric phosphorus loading, however, are not common because many studies of atmospheric inputs have been focussed on elements associated with acidic deposition such as nitrogen, sulfur, calcium, sodium and chloride (Hicks et al., 1993; Puckett, 1995). In Gorham's (1961) review of the chemistry of inland waters, phosphorus is not mentioned in the discussion of the chemistry of atmospheric precipitation. Surprisingly, even the multi-agency Great Waters Program (U.S.EPA, 1994), the National Dry Deposition Network (Hicks, et al. 1993), and the National Atmospheric Deposition Program/ National Trends Network (NADP, 1996) do not include total phosphorus monitoring from atmospheric sources, in spite of the importance of phosphorus to many freshwater systems.

Phosphorus has been shown to be limiting to algal growth in many freshwater lakes (e.g., Nichols and Dillon, 1978; Paerl, 1988; and Smith, 1990), and phosphorus control is often the primary focus of lake management (Schindler et al., 1978; Stockner and Shortreed, 1985; Welch and Cooke, 1987). In shallow, freshwater environments such as the Florida Everglades, atmospheric inputs of phosphorus are expected to be very important, because the dominant source of water for this marsh system is local convective storms (Davis, 1994). Unfortunately, there is little consistency in estimates of deposition rates for total phosphorus in South Florida. Irwin and Kirkland (1980) reported a wide range of values in their review of the historical data on precipitation quality (bulk) and concluded that these data were not sufficient to define baseline conditions for the region. Hendry et al. (1981)

also reported significant variation in P deposition among sampling sites, with values ranging from 17 to 111 mg P/m^2/yr. They found the overall state average deposition rate as bulk precipitation to be 50 mg P/m^2/yr. More recently, Dixon et al. (1996) reported annual station averages ranging from 52 to 176 mg P/m^2/yr for bulk deposition in the Tampa Bay area, with a 7-station average of 93. Peters and Reese (1995) also found data variability to be very high (e.g., wet values from 0.001 to 0.620 mg P/L) in dry and bulk bucket-type collectors near Lake Okeechobee, Florida. Such variability is not unique to Florida and published deposition rates for phosphorus outside the State range from less than 10 to well over 100 mg P/m^2/yr (e.g., Graham and Duce, 1979; Likens and Bormann, 1995).

There are several plausible explanations for this variability. For wet deposition, rainfall volume per event, interval to last event, season, and storm path can alter mass deposited in an event (Jordan et al., 1995). Dry deposition is controlled by fundamentally different physical, chemical and biological processes than wet deposition. Dry deposition can be affected by properties of the collecting surface, canopy structure, and season (Wu et al., 1994), as well as wind speed, time of day and size spectrum of depositing particles (Davidson et al., 1982; Noll et al., 1988). Another key factor is sample contamination from natural debris or that associated with birds or insects attracted to the collector device (e.g., Peters and Reese, 1995). In addition, the method of sample collection and local land use can affect absolute amounts and variability in the data (Hendry et al., 1981; Lovett, 1994). As a result of these factors and others associated with atmospheric turbulence and boundary layer phenomena, dry rates can be expected to be more variable than those for wet deposition (Hicks et al., 1993; Lovett, 1994; van Ek and Draaijers, 1994).

The lack of precision and accuracy of deposition data in general, and of phosphorus data in particular, presents an immediate challenge for environmental management. For example, the vast majority of the remnant Florida Everglades marshes are very shallow and often display ambient phosphorus levels at or below 10 µg P/L (e.g., McCormick, et al., 1996). Therefore, atmospheric loading, even at the low end of the range of published values, could be a highly significant source of enrichment to this ecosystem. Furthermore, the Everglades Forever Act passed by the Florida Legislature in 1994, requires the South Florida Water Management District to restore the remaining Everglades ecosystem, in cooperation with several other State and Federal agencies. The control of phosphorus loading to this oligotrophic, subtropical environment is a cornerstone of the restoration. Based on the literature reviewed for this study and data collected to date by various organizations in South Florida, a defensible estimate of P loading from the atmosphere with associated uncertainty

does not exist at this time for any location or for the region as a whole. This lack of definitive information on phosphorus inputs from the atmosphere is typical for most aquatic ecosystems and provides the motivation for this study. While better estimates of deposition for managing the Everglades ecosystem is a primary objective, virtually all information on phosphorus deposition and its measurement is of equal relevance to any ecosystem for which P is the target of management efforts.

The first objective of this paper is to provide a conceptual framework for information on phosphorus deposition in rainfall and dryfall from the atmosphere, based on information available in the open literature. The second objective is to summarize P deposition rates from the scientific literature in light of the conceptual framework and to evaluate these estimates for applicability to environmental management in general, using South Florida as an example. The final objective is to recommend a program of field sampling, experimentation and data analysis to improve estimates of atmospheric loading of phosphorus to aquatic ecosystems. The information analyzed to meet these objectives should be useful to environmental managers as they seek to understand the sources and sinks of phosphorus in systems where the nutrient enrichment is a target of environmental management.

Conceptual Basis for P Deposition from the Atmosphere

Components and Size Fractions of P Deposition and Associated Processes

Atmospheric deposition can be separated into 3 components for simplicity in the following analysis of relevant concepts: wet, dry and cloud deposition (Lovett, 1994). Wet deposition is that associated with directly falling atmospheric precipitation containing materials scavenged from the atmosphere as either gases or particles. The separation between dry and wet deposition is defined operationally by the typical bucket collection system that opens the wet bucket only when rain is falling in significant amounts. Deposition from clouds and fog may be difficult to separate into wet and dry components; on a practical basis, however, it is measured by dry techniques. Hicks et al. (1993) provide a good working definition of dry deposition: 'As currently defined, dry deposition is the gravitational settling of large particles not associated with falling precipitation, and the turbulent exchange of small particles and trace gases to the surface'. This definition implies that variations in dry deposition are under the control of very different physical, chemical and biological factors than is the case for wet deposition. Wet deposition can be expected to display the random variability

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associated with individual rainfall events, spatial patterns of precipitation and processes involved in scavenging of materials from the air. For example, Jordan et al. (1995) explained much of the temporal variation in wet deposition by considering season, rainfall volume, and elapsed time since last event in a regression model of a long-term data set in the Chesapeake Bay area. Dry deposition rates are determined by wholly different mechanisms from wet deposition, including particle size and concentration in the atmosphere, settling velocity and most importantly, the nature of the collecting surface. Dry deposition is fundamentally more complex and variable than wet (e.g., Van Ek and Draaijers, 1994), and will therefore be given greater attention in the following discussion. Finally, cloud water deposition occurs when fogs or clouds contact surfaces and is important in elevated coastal or mountainous environments. Although fogs do occur sporadically across the south Florida landscape, it is unlikely that fog/cloud deposition will be a significant part of long-term phosphorus input to this low-elevation subtropical environment. Cloud water deposition will not be considered further in this analysis as a distinct process. Any approach to sampling the dry and wet components will be assumed to capture materials conveyed by clouds and fog. Also, dew will be considered as a factor altering dry deposition through surface wetting (as suggested by Lovett, 1994) rather than as a separate form of deposition.

In atmospheric dry deposition, phosphorus is associated with particles, particularly those greater than 2 µm in diameter (e.g., Graham and Duce, 1982; Lawson and Winchester, 1979). In one case study (Como Creek watershed, Colorado) it was found that even after being in aqueous suspension for part or all of a 7-day sampling cycle, over 75 % of deposited P was still associated with particles (Grant and Lewis, 1982). Results such as these imply that to understand P dry deposition, and how to measure it effectively, one must consider the nature and behavior of atmospheric particles of different size classes in forming a conceptual framework for atmospheric deposition. The following discussion of concepts is organized around three size fractions of particles and summarized in Table 1.

Phosphorus-bearing particles in deposition from the atmosphere are derived largely from soil, manufacturing, dust, pollen, fuel combustion, insects, bird excrement and other biological debris, fertilizers, coal and other ash, some pesticides and aerosols from the ocean (Graham and Duce, 1979). Fine particles (<2 µm) have the potential to travel long distances in association with stratospheric or tropospheric air masses and may have residence times in the atmosphere ranging from weeks to months depending on mixing and scavenging processes (e.g., Waser and Bacon, 1995). Delumyea and Petel (1979) found that phosphorus-containing particles over Lake Huron,

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Table 1. The characteristics of atmospheric dry deposition of phosphorus. The information summarized in this table is drawn from various sources in the literature, particularly from Erisman et al. (1994), Hicks et al. (1993), Lovett (1994), and Noll and Fang (1989).

Characteristic	Particle Size Category		
Key Attributes of Dry P Deposition	Fine Aerosols and Gases (< 2 μm)	Intermediate Particles (2μm- 10μm)	Coarse Particles (>10 μm)
Source	Global/Regional	Regional/Local	Local
Land Use Association	Low density, low landscape disturbance	High density urban, or high disturbance agriculture	Dependent on local groundcover and biological communities
P Content	Low	High (0.1-0.2%)	High, variable
Transport Mechanism	Atmospheric circulation, follow local air eddies	Local and atmospheric processes, less dependent on eddies	High energy events, gravitational effects dominate settling
Settling Rate	Low (< 1cm s^{-1})	High (1-10 cm s^{-1})	Very high (>10 cm s^{-1})
Surface Effects	Fine structure and wetness are important,	Variable effects, wetness important, inertia involved	Surface less involved, inertia and sediment-ation dominate
Canopy Effects	Important as sink	Variable, resuspension and bounce-off	Important as source, esp. organic debris
Deposition Rate Variability	Lower variability, more continuous	High variation, event related	Very high, more episodic and seasonal variation

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and many miles from their continental sources, averaged about 1 μm in mass median diameter (midpoint of mass/particle size distribution), 28 % of particles were less than 0.5 μm, and between 5 and 30 % were greater than 4.6 μm is diameter. From these data, the authors concluded that P-bearing particles deposited onto Lake Huron were derived from continental erosion. Coarse particles (>10 μm as a working definition for this study) have high settling rates and may often be local in origin. However, Davidson et al., (1985 a&b), Holsen et al., (1993), Lin et al. (1994), and Noll and Fang (1989) reported that dry deposition is conveyed primarily by particles between 10 and 100 μm, well above the particle sizes measured effectively by monitoring studies designed to quantify gaseous and aerosol deposition. Coe and Lindberg (1987), Cole et al., (1990), Lawson and Winchester (1979), Lee and Patterson (1969) and Noll et al. (1988) also found that the vast majority of dry deposition was associated with particles greater than one micron in diameter. These larger particles may be present in the atmosphere at low concentrations, but have high sedimentation rates and high P content. The P content of particles can be substantial at about 0.1 – 0.2 % (Graham and Duce, 1979; Grant and Lewis, 1982), and high winds or atmospheric convection can vector large particles over great distances (e.g., Hicks, et al., 1993). Land use practices associated with agriculture or urbanization are associated with increases in suspended particles and resultant high rates of P movement and deposition (e.g., Eisenreich et al., 1977; Graham and Duce, 1979; Hendry et al., 1981; Lee and Patterson, 1969; and Noll et al., 1988).

Ecological studies have reported that particulate debris up to 1 mm in diameter can represent an important source of depositional P (Cole et al., 1990; Jassby et al., 1994; Likens, 1985). Measurement of this component is fraught with uncertainties, however. Natural detrital or dust particles in the air are difficult to separate from sample contamination by bird excrement, insects or organic matter from the area immediately surrounding the sampler; such contamination must be excluded from deposition flux. The objective for measuring P deposition for environmental management is to estimate net input per unit area as part of an overall mass balance of P in a managed ecosystem. Studies dating from the 1950s suggest that natural organic matter from the air may be a very important source of carbon or other elements (Gasith and Hasler, 1976; Gorham, 1961). The dilemma is how to decide what particulate matter is a natural part of atmospheric deposition and what is caused by contamination of the sampler or local, non-atmospheric inputs. Some sampling programs consistently preclude all larger materials from samples: Anttila et al. (1995) and van Ek and Draaijers (1994) used filtration to remove debris from samples; Brown and Lund (1994) used cotton in funnels to keep larger materials out of samples; and Schindler et al.

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(1976) placed 1.5 mm nylon screening in the bottom of collectors to eliminate large particles. To avoid local influences, Grant and Lewis (1982) covered bulk collectors with nylon screen and mounted the collectors on 4 meter towers. Jordan et al. (1995) found that pollen was a frequent component of samples and was associated with fluctuations in nutrient levels. However, these same authors discarded samples containing debris other than pollen, such as insects or other organic debris (Thomas Jordan, pers. com.). The censoring of samples that have been compromised by contamination is very common (e.g., Dixon et al., 1996), but seldom acknowledged explicitly in published data. However, censoring samples that contain visible organic matter or using other physical means to avoid larger particles in samples precludes representative estimates of deposition derived from coarse particles. This practice may result in a systematic underestimation of real deposition rates, particularly for small lake ecosystems in forested watersheds. In fact, there are estimates in the literature of substantial deposition from airborne particles greater than 1 mm in diameter, known as large particulate organic matter in ecological jargon (e.g., Psenner, 1984; Rau, 1976).

Surface effects can be very important to the deposition rates measured by various methods. Both Cole et al. (1990) and Jassby et al. (1994) used wet vessels to estimate dry deposition to a lake surface and found that the wetted surface captured much more P than the same containers without water. Both authors indicated that P was primarily in the form of coarse particulate matter. Collections during the summer in subalpine Colorado resulted in much higher deposition rates for sodium, potassium, calcium and chlorides with a wetted surface; rates of P deposition were not significantly different (Lewis, 1983). These findings are somewhat difficult to interpret, however, because the wet and dry collectors were not identical in structure. The 'wet' collector was a square plexiglass box over one-half full of water, while the 'dry' collector was an inverted pyramid connected to a funnel. Hicks et al. (1993) argued that the open water surface does not necessarily trap particles as efficiently as might be suggested by data from containers. Air flow over a wetland or lake water surface may tend to be stratified during midday when dry deposition rates are highest, and although exchange with the air may be greater at night due to convective mixing, the air layer that is cleansed of particles may be quite shallow. Clearly, more direct measurements of deposition onto open water environments are needed if we are to predict deposition rates for very large water surfaces, such as Florida's Lake Okeechobee and the Everglades.

Davidson et al. (1982) stressed the importance of the collecting surface and noted earlier work suggesting that deposition rates can vary over several orders of magnitude based on fine

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structure or geometry of the collecting surface. Thus, it would not be surprising to find significant differences in dry deposition rates in the same area within different plant canopies. The canopies tend to act as a sink for small and intermediate-sized particles (Lindberg and Lovett, 1983), but may actually be a source of larger, coarse materials. However, vegetation processing within a canopy can alter the apparent deposition rate either through uptake or leaching.

There are edge effects in forest stands so that deposition rates within 5 stand-heights of the edge are higher than those in the interior of the stand (reviewed by Erisman et al., 1994). Weathers et al. (1995) reported that cloud deposition was reduced by 50% at 28 m from forest edges. Clearly, landscape patchiness needs to be considered in determining deposition rates to ecosystems with a mosaic of plant canopies. As one moves away from a canopy source, the deposition of plant-generated materials can be expected to decline greatly with distance from the stand edge (e.g., Cole et al., 1990; Gasith and Hasler, 1976; Psenner, 1984: Richerson et al., 1970). This same trend can be seen in P concentrations in the atmosphere moving away from the coast; levels near Bermuda, about 1000 km off shore, are approximately an order of magnitude less than P concentrations at the North American coastline (Graham and Duce, 1982). For southern Lake Huron, Delumyea and Petel (1979) found across-lake gradients in P-bearing particles when the wind was moving offshore and used these gradients in concentration to calculate deposition velocities for phosphorus. The challenge is how to estimate net deposition to large and complex natural surfaces, such as the Florida Everglades. The Leaf Area Index has been found to be one quantitative means of scaling through all the potential canopy effects to arrive at a landscape level estimate of deposition (Lindberg et al., 1988). Scaling factors need to be considered when generating net P inputs from data collected by various artificial sampling regimes; the relative importance of canopy effects will vary greatly across ecosystems.

When all these factors are combined (Table 1, bottom), the net effect is that very small particles tend to deposit gradually and more continually than larger fractions. Fine particle deposition is not easily quantified, however, and estimates of deposition velocities for particles less than 1 μm have been found to vary over 2 orders of magnitude (Sievering, 1984). Measuring the fine particle component of deposition, therefore, would require continual, long-term monitoring with the means of capturing fine particles quantitatively. It is likely that this component of deposition is a minor contributor to net P flux, due to very low mass per particle. As a result from the perspective of environmental management, the submicron component of P deposition should probably not be given priority attention for quantification. The coarse particle fluxes are more

variable and event-related since high settling rates require greater atmospheric turbulence to keep particles in suspension. Holsen et al. (1993) concluded that Lake Michigan got most of its dry deposition during periods of strong southerly winds from the urban areas to the south of the Lake. Lin et al. (1994) provided direct evidence of high deposition rates associated with strong winds. Coarse particulate matter will generate variability in deposition rates stemming from transport during windy, high energy events and/or pulses of biological production such as pollen release, insect emergence, leaf abscission, epidemics of plant disease or other natural occurrences affecting organic matter distribution. Capturing the immediate and coarse fractions (Table 1) will require very different sampling strategies capable of capturing rapidly settling particles while minimizing the bias associated with artificial surfaces and contamination.

P Deposition: Integration from an Ecological Perspective

Figure 1 helps to integrate these concepts of sources, sinks and processes across the landscape, in this case patterned after South Florida. This schematic also introduces some hypothesized relationships based on another important concept for atmospheric deposition: resource spiraling. In the ecological literature, the spiraling of materials downstream through stream ecosystems has been studied in some depth (see Elwood et al., 1983 and Newbold et al., 1981). This research has provided a synthesis that links the concepts of nutrient cycling and unidirectional mass transport through stream ecosystems. In essence, matter spirals down the riverine system in alternating periods of movement and retention. The nature of spiraling for a particular element or form of organic matter is related to structural and functional aspects of the stream ecosystem.

Several studies of atmospheric deposition have suggested that materials also spiral across terrestrial systems with the atmosphere as the agent of transport (Davidson et al., 1985b; Steen, 1986). To the extent that this phenomenon does occur in atmospheric transport, it is expected to be much more irregular, without the unidirectional certainty that is inherent in flowing water systems. The process may involve simply 'rebounding' of larger particles (Davidson et al., 1982), or longer, more intricate involvement in local ecological processes and retention followed by resuspension; this may occur routinely in larger organic debris. Both Cole et al. (1990) and Jassby et al. (1995) found that surface P inputs to lakes were predominantly of local origin as coarse particles or organic matter, and must therefore be tied to some extent to local ecosystem processes. Although the vast majority of studies in atmospheric deposition view coarse particles as contaminants, from the
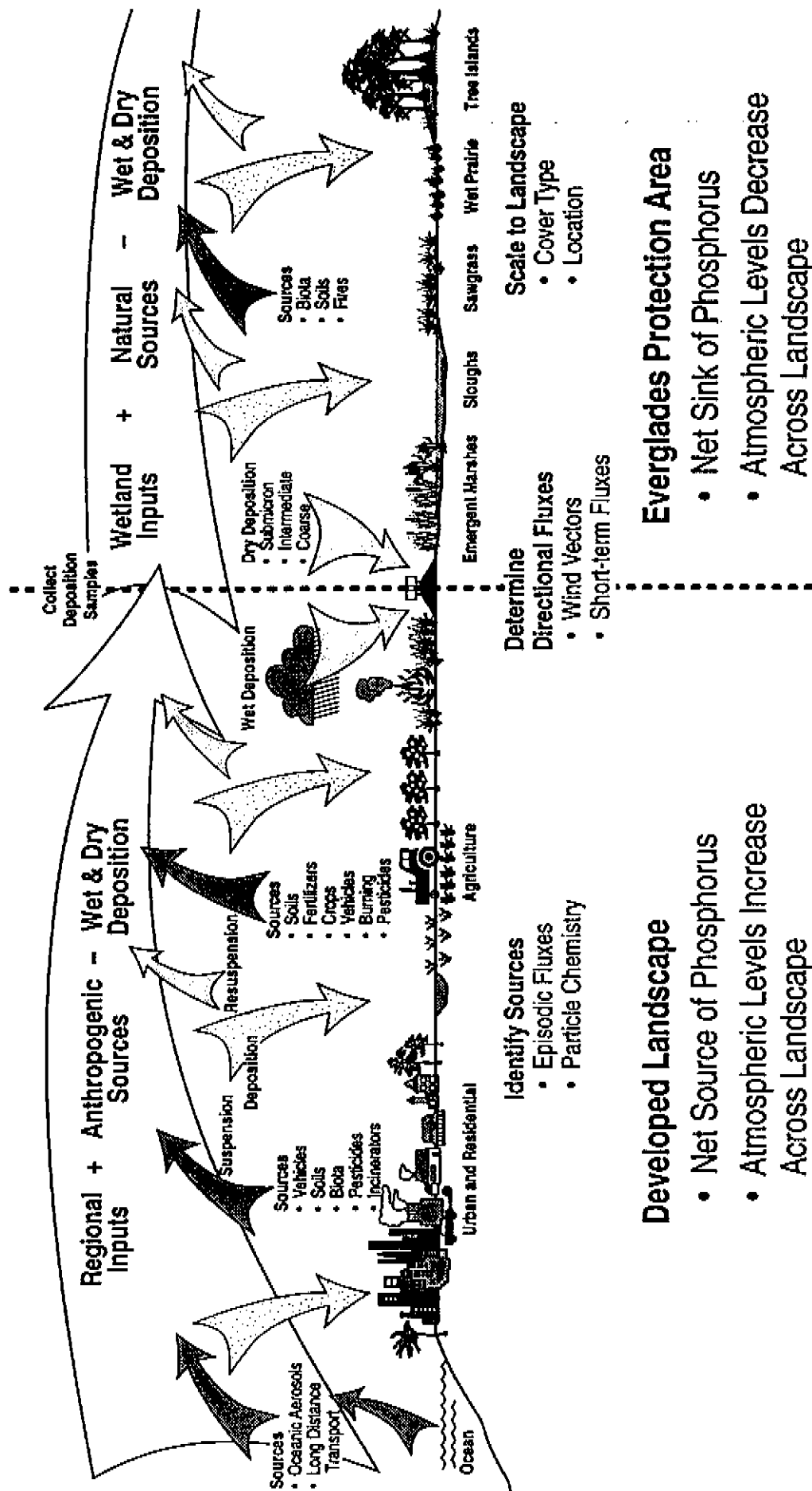
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Figure 1. A schematic representation of concepts involved in the deposition of phosphorus from the atmosphere. The landscape of South Florida is used in the illustration, although all sources, processes and other concepts can be applied to any ecosystem. The schematic also includes notes on the kinds of information needed to assess P inputs to a large aquatic ecosystem, such as the Florida Everglades.

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viewpoint of ecosystem mass balance this component can not be ignored. Coarse particulate matter may be particularly important in low-nutrient, natural environments like the Florida Everglades where even small net inputs from adjacent urban or agricultural areas may represent a significant source of P, at least over the long-term.

The factors summarized in Figure 1 produce daily and seasonal differences in deposition rates. Obviously, the timing of rainfall events will dictate when wet deposition occurs. Convective storms are most common in the afternoon and evenings from June through October over large areas of North America. Longer lasting frontal storms tend to occur in the fall and winter, and may bring with them strong winds carrying terrestrial or marine particulates for deposition. Using a network of sites across the continental U.S., Meyers et al. (1991) found that total sulfur deposition rates were highest in the summer even though atmospheric SO_2 concentrations were highest in the winter. Eisenreich et al. (1977) (Lake Michigan area, U.S.A.) and Pratt et al. (1996) (Minnesota) also found rates highest in the summer. Shaw et al. (1989) found that nitrogen and phosphorus deposition rates were highest in May and remained relatively high during the summer months in Central Alberta, Canada. They also noted that monthly P loading from the atmosphere did not correlate closely to precipitation; nitrogen loading corresponded more closely to rainfall amounts. It is also important to mention that Shaw et al. (1989) found daily P loading from the atmosphere to range over 2 orders of magnitude. Jordan et al. (1995) reported that both nitrogen and phosphorus deposition rates were highest in the spring, mimicking the pattern of precipitation. Hicks (1986), Noll et al. (1988) and Pratt et al. (1996) all reported that dry deposition rates were higher in the daytime in association with stronger winds and larger mean particle sizes.

Based on the foregoing information, studies to quantify the rate of wet P deposition from the atmosphere must account for the temporal variability of rainfall quantity and water chemistry. Studies must also be designed to convert data from a small group of individual stations collecting wet and dry fallout into deposition estimates suitable for regional application. Deposition data will need to be extrapolated over large areas, using rain data from a network of rain gauges that spans the spatial variability of precipitation and empirical relationships for areal differences in dry deposition across the landscape. For dry P deposition, studies must provide estimates of inputs (seasonal and annual) for the size fractions of depositing particles, with emphasis on intermediate sized and coarse particulate fractions (Table 1). Measurement of dry deposition must incorporate variability generated by seasonal and weather related dynamics, particularly rare, windy atmospheric events that may be associated with high deposition rates from long distance transport

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of particles greater than 10 μm in diameter. Estimates of dry deposition must also be scaled to the landscape level to account for differences in deposition associated with surface and canopy effects and with changes in atmospheric particle concentrations across the landscape. Objective, standardized procedures are needed to separate natural organic inputs of P from sample contamination, and directional fluxes should be determined to assess how closely the data on deposition rates reflect actual 'net' inputs to the target ecosystem.

Published Rates for P Deposition from the Atmosphere

The second objective of this study was to summarize P deposition rates from the scientific literature in light of the conceptual framework and to assess these estimates for applicability to environmental management in general and to South Florida, specifically. There are probably over one hundred published values for P deposition in the literature. The purpose of this investigation was not to catalogue all these values, but rather to provide an overview of annual rates to bracket long-term values that can be expected commonly in managed ecosystems surrounded by various land-uses. Several authors have invested considerable effort in summarizing earlier studies on P deposition (Ahl, 1980; Eisenreich, et al. 1977; Graham and Duce, 1979; and Likens and Bormann, 1995). The literature used in their compilations is included here as average values and ranges.

Atmospheric P concentrations set the context for summarizing deposition rates. Graham and Duce (1979) reported that P concentrations in the atmosphere over cities range from 30 to 1450 ng P/m^3, with an average of about 150 ng P/m^3, while values in coastal environments tend to range from 10 to 20 ng P/m^3. They also found oceanic values to be highly variable; higher values (ca. 5 – 55 ng) near continental sources and very low concentrations, sometimes below 1 ng/m^3, in the open marine environment. These authors also reported low P levels of 1.3 ng and 0.2 ng for remote continental areas, the Northwest Territories, Canada, and the South Pole, respectively. For 33 samples from the western North Atlantic, Graham and Duce (1982) found values ranging from 0.6 to 21.9, with an average of 6.4 ng P/m^3. In essence, concentration data tends to follow expectations based on particle sources and transport; higher values in developed areas and low concentrations in undisturbed, remote environments.

The vast majority of published rates of P deposition are estimates from 'bulk' measurements in which the sampler is left exposed to both wet and dry deposition for extended periods, usually 1 to 2 weeks. Atmospheric scientists are critical of such bulk estimates because the data are often influenced to varying degrees by local contamination due to extended periods of exposure, and

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contain some unknown fraction of dry deposition due of the geometry and aerodynamics of the collector. These considerations lead Hicks et al. (1993) to conclude: " These problems make the use of bulk deposition data prohibitively difficult." While the drawbacks of bulk collections are undeniable, the simplicity of the method and a general lack of practical alternatives encourage many investigators to continue publishing bulk deposition data. As will be discussed below, the published bulk values for P deposition are, for the most part, in reasonable agreement with data derived from separate wet and dry sampling programs, and are often consistent in overall range and average magnitude. Therefore, until more definitive estimates of P deposition are published routinely, we must continue to rely on historical bulk deposition data to assess atmospheric inputs of phosphorus for environmental management. This decision is purely pragmatic for the purposes of this study, however, and should not be interpreted as an endorsement of bulk measurements for new deposition sampling.

The results of the literature review of annual mean rates of P deposition, in ascending order, is provided in Table 2. Although the range of published values in the cited references in Table 2 is from 3.6 to 200 mg $P/m^2/yr$, most published deposition rates fall between 20 and 80 mg $P/m^2/yr$. In marine environments, deposition tends to occur at low rates (Graham and Duce, 1979), in spite of the fact that the surface microlayer is enriched with P relative to the underlying water, and can be a source of atmospheric P (Graham et al., 1979). In Florida, Hendry et al. (1981) reported a low deposition rate of about 17 mg $P/m^2/yr$ for a maritime station in the Florida Keys. Inland sites associated with northern forests of the midwestern U.S. or Canada tend also to have rates of deposition less than about 20 mg $P/m^2/yr$. Rates in the middle of the range reflected in Table 2 are from about 30 to 50 mg $P/m^2/yr$, and provide a first approximation of regional rates for continental U.S. Values of 50 or more are associated with studies from highly developed areas in Western Europe and the U.S. with agricultural and urban land uses. The mean ratio of Total Nitrogen : Total Phosphorus (TN:TP) in annual values of Table 2 is 25, and there is a significant inverse association between P deposition rates and the TN:TP ratio in deposition. The nonparametric Spearman rank correlation coefficient between P deposition rates and TN:TP ratios in Table 2 is highly significant (r, = 0.90, n = 13); thus, P limitation of primary productivity will tend to be maintained or enhanced by atmospheric nutrient inputs in less disturbed, forested landscapes. Jassby et al. (1994) concluded that the N:P relationship in atmospheric deposition to Lake Tahoe (CA/NV) was consistent with the observed shift towards P limitation in this oligotrophic, subalpine lake.

The information in Table 2 provides estimates of deposition useful for assessing the relative

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Table 2. A summary of annual mean rates of phosphorus deposition from the atmosphere into terrestrial and aquatic ecosystems. Deposition values are from published measurements ranging from about one to over twenty years. These data have not been selected for any particular ecosystem type, and are presented to illustrate typical rates on an annual basis. Note that several of the values are averages from many studies reviewed in the cited articles.

P Deposition Rate, mgP/m²/yr	N Deposition Rate, mgN/m²/yr	Sampling Location and Cover Type	Notes on Sampling Design and Techniques; Misc. Comments	References
5.8 (w/o 1 high value)	None	Bermuda; marine	On shore at Bermuda, single bulk collector exposed 24 to 96 hrs. 14 samples, 1974	Graham and Duce (1982)
7.0 (1 year)	None	Narragansett, Rhode Island; coastal urban	Concentrations in precipitation samples, bulk collections with rain events	Graham and Duce (1979)
12.4 (6 years)	795	Shagawa Lake, Minnesota; forested, small town setting	Bulk conc. in precipitation samples at city of Ely, Minn.; Bulk conc. X monthly precip. on lake; range 8.8 – 17.6 mg P	Malueg, et al. (1975)
14. (5 years)	490	Northern Finland, misc. forested	Bulk deposition (assumed), 8 stations, 5 year period, 1971-1976; interpreted as background rate; range 6 – 22 mg P	Ahl (1980) (Table 1)
20.3 (3 years)	424	Narrow Lake, Alberta, Can.; forested	3 to 5 bulk collectors, sampled after events and dry periods, funnels on collectors	Shaw et al. (1989)
23.8 (16 years)	1103	Lakes in Central Ontario, Canada; forested	Mul. & var. sites, 1976 - 1992, bulk collections, sampling median 8 days, w. window screens; range 11.1 – 74.9 mg P	Reid and Dillon (1994)
23.8 (20+ years)	749	Mean for 17 forests in U.S. and Western Europe	Various annual mass balance studies of key elements, primarily bulk collections; range 3.6 – 60 mg P	Likens and Bormann (1995) (Table 24)
27.8 (1.5 years)	None	Lake Michigan area; urban settings	Data from ref. Figure 2, 15 sites sampled monthly for 17 months, bulk collections	Eisenreich, Emmling and Beeton (1977)
30. (mul. years)	None	North America continental average (Table 6)	25 authors summarized in ref., various approaches, 1960 to 1976; range 7 – 150 mg P	Graham and Duce (1979) (Table 3)
30.7 (1 year)	None	Lake O'Woods, W. Virginia.; forested	Single collector, sampled after rainfall events, bulk collections, large particles screened	Havens and DeCosta (1984)
32.6 (4 years)	562.1	Lake Tahoe, Ca/Nv, 1900 m elevation; forest & urban setting	Ward Valley Lake Level station, 1989-1992, wet and dry bucket collections; dry + distilled water	Jassby et al. (1994)

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P Deposition Rate, mgP/m²/yr	N Deposition Rate, mgN/m²/yr	Sampling Location and Cover Type	Notes on Sampling Design and Techniques; Misc. Comments	References
32.5 (4 years)	624.6	Rawson Lake, Ontario, Can.; forested	Single station on island in lake, 4 years of data, bulk collections with 1.5mm screening; range 24 – 53 mg P	Schindler et al. (1976)
33.8 (mul. years)	None	Great Lakes, regional mean	9 annual means, 6 studies, bulk samples, monthly data; range 8.9 – 102 mg P	Eisenreich et al. (1977)
35.1 (1.5 years)	None	North Yorkshire, England; rural near Barlow	Barlow Station deposition as dust collection; monthly, 17 months; data from Frisbee w. foam insert (64.1 mg dust/m2/dy X 0.0015 P/mg)	Vallack (1995)
35.8 (1 year)	587.6	Piburger See, Austria; forest	5 stations on lake for large material + 10 for fine; biweekly, bulk samples; large samplers + 1mm screen, small w/o	Psenner (1984)
41.0 (12 years)	1176	Chesapeake Bay; rural, forested, wetland;	Single site, event sampling since 1978 for TN and 1980 for TP, bulk collections w. 28cm funnel, 13 m tower; Aerochem for wet only; range 27.4 – 70.6 mg P	Jordan et al. (1995) (Balti./Wash. Airshed)
50. (mul. years)	None	Western Europe, regional ave.	16 authors reviewed in ref. Table 4, ave. in ref. Table 6; Various approaches, 1951 to 1976; range, bulk data 20 – 200 mg P	Graham and Duce (1979) (Table 6)
50 (1 year)	750	Florida, coastal, rural,agric. and urban	24 sites within the State, sampled biweekly, bulk collections using buckets for 12 months in 1978/79; range 17 - 96	Hendry et al. (1981)
59 (1 year)	910	Florida,urban, maritime, rural and agricul	Sites at 4 locations in Florida sampled biweekly, wet/dry buckets, Aerochems, 12 months 1978/79; range 24 – 96 mg P	Hendry et al. (1981)
60.5 (1 year)	None	Lake Carl Blackwell, OK.; agricultural	3 stations, 13 periods; post event sampling, bulk collections with #18 wire screen (data variable)	Sober and Bates (1979)
93.3 (1 year)	859	Tampa Bay watershed; misc. landuses	7 stations, sampled biweekly Bulk collections using funnel samplers; value is mean of 7 stations; range 52-176	Dixon et al. (1996)
126.6 (1 year)	957.4	Dunham Pond, Conn; forest and wetlands	Single station, concentrations and precipitation, bulk collections, sampled biweekly (N.Y. city airshed)	Kortmann (1980)

importance of atmospheric inputs into ecosystems, including those in South Florida. We should expect values less than 20 mg $P/m^2/yr$ for sparsely populated areas near the Florida coast. Inland rates will vary with land use. High P inputs (60 to 100 + mg $P/m^2/yr$) can be anticipated from urban or agricultural areas; Hendry et al. (1981) reported high values for urban (48 mg $P/m^2/yr$) and agricultural (66 mg $P/m^2/yr$) areas of Florida. An overall rate for the remnant Everglades marshes will probably lie between 30 and 40 mg $P/m^2/yr$ (McDowell et al., 1997). This range of values is consistent with some P accumulation rates for sediment cores taken in South Florida, published values for relatively undisturbed areas, and data from Florida studies of deposition rates.

Quantifying the Atmospheric Deposition of P for Ecosystem Management

Are such approximations from published values defensible for decision-making in ecosystem management, or are additional measurements warranted? Obviously, each manager will have to assess the potential importance of atmospheric inputs in order to decide on how much technical effort should be given to reducing the uncertainty of P inputs from the atmosphere. In oligotrophic systems with low hydraulic loading rates, atmospheric influences can be expected to be more important and may be effected greatly by land use changes. Regarding South Florida, the majority of P loading to the pristine portions of the Everglades ecosystem comes directly from the atmosphere, and the remnant system is, as a whole, very oligotrophic. In fact, there is evidence that the periphyton assemblage of the Everglades can be altered at P levels between 10 and 20 µg/L (McCormick et al., 1996). With these facts in mind, it is clear that more accurate and precise information is needed for South Florida than can be derived from earlier studies of atmospheric deposition.

Once it is determined that more information is needed, the next step is to decide how to collect the data. Simple 'bulk' precipitation measurements provide background, but, as mentioned earlier, the drawbacks of this approach render it inappropriate for modern studies. The dilemma for environmental management is that atmospheric science over the last several decades has been focussed on measuring acid precipitation, and many of the extant monitoring programs reflect an interest in nitrogen, sulfur, ozone and other constituents associated with acidic inputs to ecosystems (Hicks et al., 1993). Unfortunately, this emphasis has produced techniques and expertise primarily on gases or aerosols. The analysis that follows attempts to guide the environmental manager through available methods to arrive at recommendations on how to improve estimates of P

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deposition rates from the atmosphere, particularly for the dry component. The evaluation is organized around the recommendations of Hicks et al. (1993), Erisman et al. (1994), Lovett (1994) and others that wet and dry deposition must be monitored separately, and that an estimate of total P deposition will be most defensible if approached using several techniques simultaneously.

Wet Deposition of Phosphorus

At the present time, automatic deposition collectors manufactured by Aerochem Metrics[*], Inc are used commonly for measuring wet and sometimes dry deposition. These samplers consist of a wet and dry bucket, each 29.4 cm in diameter. The wet bucket is uncovered only during periods of rainfall, while the dry bucket is left open all the time except for periods of active precipitation. For wet deposition, the Aerochem collector appears to represent an adequate sampler. In fact, the National Atmospheric Deposition Program (NADP) recommends the Aerochem Metrics 301 Collector for its nationwide network of wet deposition sites, and the device has been used widely in other studies of wet deposition (Hendry et al. 1981; Jassby et al., 1994; Jordan et al., 1995; Landing et al., 1997; and Peters and Reese, 1995). The wet bucket collects rainfall in amounts very close to those attained by independent rain gauges (SFWMD, unpubl. data) and there are no cases of systematic bias in these samplers for the collection of data on rainfall water quality and rainfall quantity reported in the deposition literature.

Challenges remain, however, in using Aerochem Metrics[*], Inc., collectors for wet deposition. In active wildlife areas, there will always be some contaminated samples from biological debris. Standard Operating Procedures need to be developed for field sampling and these should include criteria for site selection; McDowell et al. (1997) recommend mounting collectors on towers of about 10 m height to avoid large local particles and minimize biofouling. Also, procedures are needed for data screening to deal objectively and consistently with contaminated wet samples. Once standard procedures are in place, data can be used to assess the ability of a network of sites to provide estimates that encompass spatial variation in P deposition rates. Information is needed on the scaling of data from these collectors to a much more extensive network of rain gauges that may often be available in a region.

Dry Deposition of P-Bearing Particles

Table 3 summarizes available methods for estimating the dry deposition of phosphorus in three general approaches: micro-meteorological methods, surface accumulation methods and inferential techniques. Reviews by Erisman et al. (1994), Hicks et al. (1980 & 1993), Lovett (1994) and

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Table 3. Techniques for estimating dry P deposition from the atmosphere into terrestrial and aquatic ecosystems. This table summarizes published methods that have the potential to provide data on the rates of dry P deposition useful in environmental monitoring and assessment. Notes on applicability are included to highlight strengths and constraints of methods for estimating particle flux in nature.

Methods, Pooled by Approach	Overview of Methods and Key References	Applicability to Estimating P Deposition Rates for Environmental Monitoring and Assessment
I. Micro-Meteorological Methods Generic Aspects	Flux is calculated from air concentrations above the surface and relationships with parameters associated with turbulent exchange processes (Baldocchi et al., 1988; Hicks, 1986).	Continuous measurement of chemical concentrations in the air and meteorological parameters requires a major sampling effort. Methods are best applied to gaseous or submicron forms and no routine micro-meteorological method for particles is available (Erisman et al., 1994). Methods are most useful for short-term studies of deposition mechanisms and effects of atmospheric conditions.
A. Eddy Correlation	Vertical wind velocity and gas concentrations allow direct estimate of turbulent transport across a plane (Meyers and Baldocchi, 1993).	Rapid response instruments are needed, usually tower mounted. Air movement is measured in 3 dimensions, as are scalars such as O_3, H_2O and SO_2. Uncertain utility for P until studies find appropriate scalars for aerosol P and the need to quantify P in fine aerosol fraction is established.
B. Vertical Gradient Techniques	Air concentrations at various heights plus meteorological variables are used to estimate eddy diffusivity and deposition velocities.	Detectable gradients, bi-directional fluxes and adequate fetch are problems, and canopy structure affects diffusivities greatly (see Baldocchi et al., 1988). The need to quantify fine P particles must be established and aerodynamic gradients are probably not measurable for larger particles subject to sedimentation.
C. Eddy Accumulation	Air sampled at rates proportional to velocity for updrafts and downdrafts. Eddy flux estimated by concentration differences (Hicks, 1986).	Must sample over wide dynamic range and small differences in concentration must be detectable. May be suitable for P if air concentration differences are detectable and air samplers can operate over local range of vertical wind velocities. Attempts to apply this approach in the field have proven unsuccessful (Baldocchi et al., 1988).

D. Bowen-Ratio Technique	Energy balance and calculated fluxes provide eddy exchange coefficients for a scalar applicable to target species based only on vertical gradient (Meyers et al., 1989 and 1996).	Relationship with gradient and flux of CO_2, H_2O, or other scalar is assumed. Likely to be useful only if a scalar like aerosol SO_2 can be determined to be applicable to size fractions of particulate phosphorus and if such fractions represent a significant input of P. The energy balance approach does not appear to be applicable to coarse particles conveying P.
II. Surface Accumulation Methods Generic Aspects	Pollutant accumulation rates are estimated from mass changes directly on surfaces, natural or surrogate (Erisman et al., 1994; Nicholson, 1988).	Best for sedimenting particles (>2μm). Note that small particles accumulate on downward and upward facing surfaces. Combinations of surrogate and natural surface accumulation measurements may be appropriate for environmental monitoring; calibration to estimate deposition onto natural surfaces is needed.
A. Net Throughfall	Difference between wet deposition in an open area and under a canopy estimates net dry deposition.	Sampling reflects the sum of wet, dry, cloud and dew deposition onto natural plant communities (Lindberg et al., 1986; Brown and Lund, 1994). Must know vegetative processes to interpret data (foliar leaching or uptake). Probably not suitable for routine monitoring due to foliar processing of P and difficulties with *in situ* measurements.
B. Surrogate Surfaces, including Aerochem Metrics, Inc., wet/dry collectors	Net mass accumulation on an artificial surface is measured over specified period of time (ca. 1 to 7 days) (Davidson et al., 1985a).	Data can be useful for coarse particles, but may not simulate deposition onto complex natural surfaces. Wind losses, surface characteristics and sample contamination are concerns. Using water in dry collector increases capture efficiency and may provide better estimates for aquatic systems; collector geometry alters deposition estimates. Field trials are needed to compare and calibrate methods for use in routine monitoring of P deposition.
C. Watershed Mass Balance	Outflow mass and information on watershed sources and sinks are combined to estimate deposition by difference.	Can give an estimate of total depositional inputs. Limited to cases in which sources and sinks are quantifiable. Probably not appropriate as a routine method for assessing P deposition into managed ecosystems.
D. Surface Accumulation on Vegetation	Mass accumulation over a period of several days on leaf surface; may be in combination with surrogate samplers (Lindberg and Lovett, 1985).	Provides direct estimate of dry deposition. Canopy processing and surface geometry can interfere. Cloud/dew droplets can change rates. Foliar interactions with deposited P may be a substantial interference for estimates to a system with plant canopy. Probably not appropriate for routine monitoring of deposition.

III. Inferential Techniques Generic Aspects	Air concentration data are combined with meteorological measurements and a dry deposition transfer model to estimate depositional velocities and fluxes (Hicks, et al., 1987).	Dependent on multiple estimates of resistance with many assumptions inherent to the transfer model. Models have been calibrated extensively (Meyers and Baldocchi, 1988). Merits some consideration for routine studies, at least in combination with other methods. However, these methods are often applied to fine aerosols and may not be applicable to larger particles conveying most P into natural systems.
A. Low Volume Filter Packs and DRYDEP2 Model, developed by the NOAA Air Resources Laboratory	Air concentrations of submicron particles and gases are derived from filter pack samples, and are combined with meteorlogical parameters to yield deposition rates through canopy models (Meyers and Baldocchi, 1988; Pratt et al., 1996).	Approach appears to produce conservative estimates and does not estimate deposition of supermicron particles. Approach is being used for Tampa Bay Estuary Program and in USEPA's Dry Deposition Monitoring Program. It may prove useful monitoring fine P particles, should this fraction need to be quantified. Supermicron and coarse particles will need to be handled by other approaches. Validation of modeled parameters is a major uncertainty (Lovett, 1994).
B. Rotary Impactor and Deposition Plates	Large volumes of air are sampled by multi-stage impactor and change in mass of each stage yields size-specific concentrations. The device is used in combination with other collector types (e.g., plates) to derive depositional velocities. (Noll and Fang, 1986 & 1989; Noll et al., 1988).	Rotary device provides a distribution of mass by four to eight size classes and associated concentrations. A flat plate is used to estimate mass flux. Application to routine data collections might be appropriate in combination with other measures of fluxes. With high volume sampling, supermicron particles can be quantified and are expected to provide the majority of mass flux for phosphorus. Comparison with other methods is needed to provide a method that will measure most P flux. Approach can be used with tracers, natural or surrogate, and could be configured as upwind/downwind stations (e.g., Delumyea and Petel, 1979) for additional information on the net effects of terrain and mean deposition velocities over diverse canopies.

Nicholson (1988) provide detailed comparisons of various methods for assessing gaseous, aerosol and particulate deposition. Much of this review material need not be repeated here; Table 3 provides a summary of facts on each technique with an emphasis on cases most closely associated with particulate measurements applicable to particles conveying phosphorus and with consideration of the potential for routine quantification for environmental studies.

Micro-meteorological methods (Table 3, I.) may be useful for quantifying the submicron component of dry deposition (see Baldocchi et al., 1988). Applicability for small particles depends on appropriate scalars, such as particulate SO_2, which can be monitored successfully, or on the ability to sample particles continuously with a rapid response particle analyzer, such as the TSI model 3320 (T. Meyers and S. Lindberg, pers. com.). However, since, as discussed earlier in this paper, P deposition is conveyed mostly by particles much greater than 2 µm in diameter and effected by sedimentation, micro-meteorological methods are not well suited to measuring P deposition rates in nature (Nicholson, 1988). Eddy accumulation has promise if larger particles can be sampled isokinetically, and if upward and downward gradients can be detected routinely; field applications of this approach have so far proven to be problematic (Baldocchi, et al., 1988).

Surface accumulation studies (Table 3, II.) have proven more useful for intermediate-sized (2 – 10 µm) and coarse particles (> 10 µm) (Holsen et al. 1993; Lin et al., 1994; Davidson et al., 1985a). The net throughfall method is impractical for phosphorus, since canopy processes are likely to be an important confounding factor and difficult to quantify. There are some inherent deficiencies in surrogate surface collectors, like the common Aerochem Metrics[*], Inc. dustfall buckets, for collecting quantitative samples of dry deposition. Left exposed to the atmosphere for up to a week at a time, surrogate surfaces are subject to persistent contamination (biofouling) by bird excrement and miscellaneous debris that may or may not be considered part of natural deposition. Birds are a particular challenge to deposition measurements in marshes; any object that can be used as a perch in wetlands will be subject to unnatural levels of contamination. The geometry of dustfall buckets is also a problem, according to Hicks (1986) and Davidson et al. (1985a); dustfall buckets have a high surface to bottom area ratio of 4.8 as compared to vessels with smaller sides such as the common Petri dish with ratio of 1.8. This high surface to bottom ratio makes dry deposition data difficult to interpret as deposition per unit surface area of a canopy or ground surface. In addition, bucket geometry creates an aerodynamic environment that decreases collection efficiency. The distortion of air flow by the bucket rim causes particles to change their trajectories and escape capture by the bucket (see Dombrowski et al., 1995). Small particles (2 – 10 µm) will be collected by such vessels

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only when vectored by downdrafts, and even then, may be subject to rebounding and resuspension. For such small particles, the chemical and physical nature of the surface can also be a factor in observed mass accumulations (Dasch, 1983 and 1985). Together, these considerations make the interpretation of dry deposition data from dustfall buckets problematic (e.g., Nicholson, 1988). These collectors probably provide more reasonable estimates for the deposition rates of coarse particles (>>10 μm) with very rapid sedimentation rates, but additional field and laboratory trials are needed to determine how these devices collect intermediate sized particles and estimate the deposition onto natural canopies. Uncertainties in what the dustfall buckets actually measure has lead the National Atmospheric Deposition Program to stop requiring the processing of samples collected from the dry side buckets of Aerochem Metrics[®], Inc. collectors in the NADP nationwide deposition monitoring network. Recently, a panel of atmospheric scientists recommended discontinuing dry bucket sampling for atmospheric deposition monitoring in the Everglades ecosystem (McDowell et al., 1997). The panel concluded that, even if contamination were not a problem, data from dry bucket collections are not interpretable for estimating P deposition rates. Other approaches using accumulation on surfaces, including watershed mass balance and accumulation on vegetation, do not appear viable as practical techniques for environmental management.

Inferential methods (Table 3, III.) depend on the basic relationship that deposition is a function of air concentration and deposition velocity. Inferential techniques can provide reliable estimates for aerosols, gases and intermediate sized particles for which air concentrations can be measured and deposition velocities can be estimated by models driven by meteorological data (e.g., Hicks et al., 1991; Meyers and Baldocchi, 1988; Pratt et al., 1996). Sampling large volumes of air, such as with a rotary impactor, might allow quantification of particle concentrations in size classes above 10 μm, which may prove to contribute substantially to P dry deposition. The difficulty is getting air concentration or deposition velocity data for large particles with rapid sedimentation rates. Combining large particle accumulation data from a standardized plate with air concentrations derived from large volume samplers is likely to yield good estimates of dry deposition for phosphorus, such as has been done by Davidson et al. (1985b) and Lin et al. (1994) for other deposition parameters.

A combination of approaches in Table 3 will be needed to improve estimates of dry deposition rates for environmental management since the goal is to measure total P deposition and there is no single technique that can give all the necessary information. The only way to get total P

deposition is by comparing and combining data from several approaches. Although sampling network design should be tailored to the objectives of a particular sampling program (Hicks et al., 1993; Lovett, 1994), the primary objective of atmospheric monitoring for environmental management is to quantify net P deposition per unit area of the target ecosystem. To reach this objective, calibrated surface accumulation methods (Table 3, II) (Nicholson, 1988) will be required to sample intermediate and coarse-sized, phosphorus-bearing particles. Using plate sampling devices, such as those used by Davidson et al. (1985a), Holsen et al. (1993), Lindberg and Lovett (1985) or Noll et al. (1988), will provide direct measurements of total particle flux by size class.

Flux information from surrogate surfaces will need to be melded with size specific atmospheric concentrations. These data can be obtained with high volume aerosol samplers and/or rotary impactors (Noll et al., 1988). The application of upwind/downwind concentration gradients of P-bearing particles across the aquatic environment may also prove useful when combined with meteorological data, as shown by Delumyea and Petel (1979) for Lake Huron and Holsen et al. (1993) for Lake Michigan. This inferential, gradient approach is particularly appealing for large aquatic ecosystems, such as the Florida Everglades, because it yields direct information on concentrations and deposition velocities for several size classes of particles and should result in validated estimates of P deposition. This sampling technique can also be adapted to using natural and surrogate tracers to improve understanding of particle attribution and deposition rates (Gatz, 1986; Garland, 1986; Sehmel, 1983). A sampling approach using two sites and samplers at multiple heights was recommended for South Florida by McDowell et al. (1997) to allow the separation of local from regional particle inputs. Overall, this design follows the suggestion made by Hicks et al. (1993) to rely most heavily on inferential techniques in developing networks to characterize deposition. However, since getting solid estimates of deposition velocities and rates of total deposition are crucial, atmospheric sampling must be combined with surface accumulation studies using several collecting devices (e.g., Holsen et al., 1993).

Additional Steps to Reduce the Uncertainty in Rates of Atmospheric P Deposition

The conceptual model for deposition, annual deposition rates from the literature and approaches to deposition measurement given above, provide a framework to assess importance of atmospheric deposition and to acquire additional data as needed. Local data, either from existing bucket collectors or new studies of the suggested design, can be used when warranted to reduce uncertainty further, and may often be worthwhile in areas with highly disturbed landscapes or with

possible point sources of phosphorus (open pit mines, municipal incinerators). The following thoughts provide guidance for progressive refinement to improve estimates of atmospheric P inputs. Some of these steps are derived from recommendations of an Advisory Panel of five atmospheric scientists who participated in a conference on P deposition at the South Florida Water Management District, October 20-22, 1997 (McDowell, et al., 1997). Other recommendations have been developed strictly from the author's viewpoint in an environmental management agency with a monitoring network for deposition and existing data sets containing a relatively large proportion of contaminated samples. Although these recommendations were initially developed with the Everglades ecosystem in mind, they should be useful to environmental managers generally.

1. Improve statistical tools for data screening and analysis.

It may often be the case that data exists for atmospheric P deposition from Aerochem Metrics® Collectors, but contains contaminated samples; this is certainly the case for southern Florida. Contamination of wet samples can be reduced greatly using rigorous criteria for selecting sampling sites in locations without undue attraction to wildlife, applying detailed sample processing protocols and conducting quantifiable visual inspection of samples. Nevertheless, contamination can never be completely eliminated, particularly for dry deposition samples. For such data to be utilized, some sort of data screening is needed beyond just the application of field notes. The degree of contamination reflected in field notes correlates poorly with nutrient levels in some samples (SFWMD, unpubl. data). An idea worth testing is the use of new techniques for rapid detection of coliform bacteria in water to determine if a sample should be analyzed by the laboratory or flagged if already processed (P. McGinnes, SFWMD, pers. com.).

For both wet and dry monitoring programs, specific statistical tools are needed to hone human judgment in quality assurance to ensure that all aberrant data are appropriately flagged before analysis and interpretation. Ahn (1998a) provides a two step approach: 1) making judicious use of field notes to screen for contamination, and 2) data screening with a linear regression of Studentized residuals to identify outliers relative to a clean subset (derived statistically) of the data. Together, these steps appear to be effective at detecting outliers in deposition data sets from 15 sites in South Florida; 47 % of wet deposition data from 1992 to 1996 was reported to be contaminated using this approach to data screening.

Since flagging may remove substantial numbers of data points from sequential databases, data analysis must deal with data gaps generated by the QA/QC process when field contamination rates are high. While simple methods of interpolation can be used when data gaps are relatively

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rare, Ahn (1998b) used expectation-maximization with Kalman filtering and a smoothing algorithm in multiple time series to form the basis of statistical models to deal with data-set after extensive censoring for contamination. Preliminary results of these analyses look promising in deriving unbiased means and standard deviations for data sets from wet deposition measurements in South Florida with large amounts of censored data.

The censoring and data modeling approach of Ahn (1998 a&b) used the information contained in uncensored samples, representing only about one-half of wet deposition samples. Using data from 4 Aerochem Metrics® 301 Collectors, Walker and Jewell (1997) applied a statistical treatment to both wet and dry deposition that makes use of a larger fraction of samples, including those with some degree of contamination. After censoring the field data for gross contamination, they assigned various categories of contamination scores +1 or −1 for presence or absence of a contaminant, and developed a linear regression model of P deposition rates in the uncensored data. This model was then used to predict contamination-free P deposition rates on a weekly basis. Although this approach requires detailed observations on sample condition, it has the advantage of extracting more information from processed samples containing some amount of material beyond that expected to occur naturally.

Once data quality control is better standardized and statistical approaches are validated, existing data can be interpreted. Screened data sets may prove able to provide better estimates of local P deposition, look into aspects of spatial and temporal coverage, and begin the process of using the data for network optimization and integrated landscape-level estimates of ecosystem inputs. However, the censoring of data or statistical correction for contamination will always carry forward some degree of uncertainty from the original data and may cull out samples conveying natural variation of P deposition rates. Independent, field measurements of P deposition rates are needed in order to assess how well any statistical treatment comes to deriving realistic estimates of deposition from data sets with contaminated samples. Using multiple measurement methods simultaneously, as suggested above, can provide an opportunity to test the ability of various statistical treatments to provide total P deposition rates accurately.

2. Determine probable sources of phosphorus-bearing particles in atmospheric deposition.

There are several approaches to determining the sources of atmospheric particles suggested by the South Florida Advisory Panel (McDowell et al., 1997). Ionic ratios, isotopic ratios, size spectrum analysis and other indicators should be applied to samples from the monitoring studies to provide information on source attribution of airborne P entering the target ecosystem. For South

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Florida, particular attention is needed on the role of urban incinerators, agricultural burning and soil suspension, phosphorus and limestone mining and other possible point sources of airborne phosphorus. The air injection of phosphorus-enriched particles from the surface of the ocean or large lakes, such as Lake Okeechobee, during wind events should at least be considered with some *in situ* measurements and analysis, as done by Graham et al. (1979). Since aerosols from the surface film of large aquatic environments tend to be enriched relative to underlying waters, there is potential for substantial P transport under certain weather conditions.

Concluding Remarks

A better understanding of P deposition from the atmosphere will contribute directly to the certainty of decision-making in eutrophication management worldwide and is most urgently needed for environmental management of oligotrophic systems. Greater precision and accuracy in deposition rate estimates is fundamentally important to mathematical models of water quality, nutrient dynamics and ecosystem processes. In South Florida, as surface water nutrient loading to the Florida Everglades is reduced, atmospheric loading will retake its historical place as a primary source of nutrients to South Florida's 'River of Grass.' Hopefully, the conceptual model, literature review and derivative steps presented above will contribute to an improved predictive understanding of the transport and impact of airborne phosphorus in a restored Everglades ecosystem. An improved understanding to deposition will also contribute to wise decision-making in other areas of South Florida, such as Lake Okeechobee, the Kissimmee River and Florida Bay, where nutrient issues are an important concern for environmental management. Obviously, the need for better estimates of P movement from the atmosphere is not limited to South Florida and P deposition rates can be highly significant in many freshwater systems which tend to be sensitive to P loading. It is hoped that this study will draw attention to the need for this information and encourage data collection and publication on P deposition rates. Total phosphorus conveyed from the atmosphere deserves to be assessed routinely through national monitoring programs such as the National Atmospheric Deposition Program, National Trends Network, Great Lakes Atmospheric Deposition Network and National Dry Deposition Network.

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